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(54) PRESSURE SENSITIVE ADHESIVE COMPOSITONS

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See application file for complete search history.

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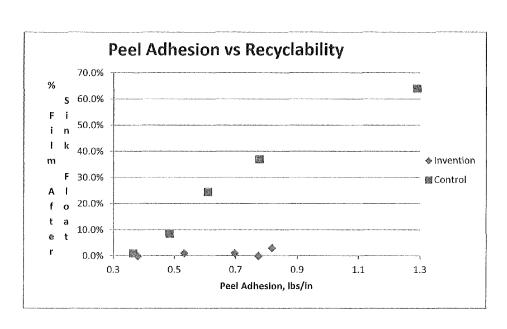
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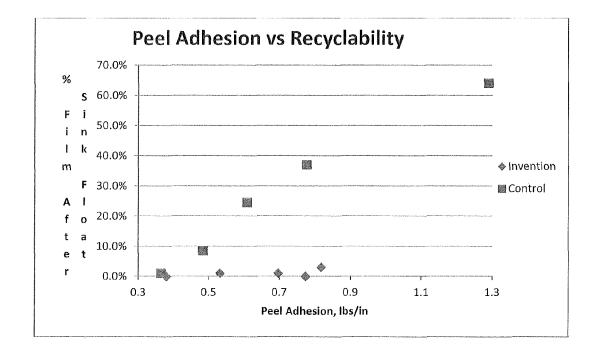
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(57) ABSTRACT

Disclosed are removable pressure sensitive adhesive compositions (PSAs) useful with facestocks and/or packaging labels designed to be removed in the recycling of polymeric packaging. The PSAs of the invention include (a) at least one monomer which may undergo free radical polymerization and whose homopolymer has a glass transition temperature of less than or equal to 10° C., (b) at least one monoolefinically unsaturated monomer having aldehyde or ketone functionality, and (c) an effective amount of at least one polyhydrazide crosslinker having hydrazine functionality. The PSA composition of the invention exhibits a peel adhesion value of between about 0.4 lbs/in (70 N/m) and about 4 lbs/in (700 N/m), and the percent film remaining on the polymeric packaging after a sink-float testing is 0% to about 8%.

20 Claims, 1 Drawing Sheet





PRESSURE SENSITIVE ADHESIVE COMPOSITONS

FIELD OF THE INVENTION

The present invention relates to aqueous emulsion pressure sensitive adhesive (PSA) compositions useful with facestocks and/or packaging labels designed to be removed in the recycling of polymeric packaging.

BACKGROUND OF THE INVENTION

In recent years, use of polymeric packaging (e.g. polyethylene terephthalate (PET), high density polyethylene (HDPE), polyvinyl chloride (PVC), polypropylene (PP), or 15 other plastic types) has increased due to their lightweight and break resistance. Pressure sensitive filmic labels have also grown in usage with polymeric packaging systems due to improved moisture resistance and appearance, and the ability to recycle such packaging materials is desirable. 20 compositions. During a typical recycling process, a hot caustic bath containing surfactant is used to release the ground plastic flake from the label. Subsequently the plastic flake is separated in a sink-float process from the removed label. The Association of Postconsumer Plastic Recyclers (APR) have established 25 protocol for evaluating labeling systems for recyclability. Failure to remove the filmic labels in the recycling process taints the color and reduces the clarity of recycled PET.

Filmic labels are more resistant to the recycling conditions which leads to a portion of the label still attached at the end of the process. Therefore, it would be desirable that the filmic label and adhesive be designed to release in the washing step of the recycling process, and to separate from the PET in a liquid, showing a specific gravity allowing for floating or sinking of the separated components.

Accordingly, there is a need for a pressure sensitive adhesive (PSA) for filmic labels that can facilitate separation in the recycling process while maintaining the desired performance properties through the article's life cycle.

There is also a need in the art for polymeric packaging 40 systems that can provide for a recycling process which is efficient and cost effective.

SUMMARY OF THE INVENTION

In one embodiment, there is provided a filmic facestock material for polymeric packaging which includes a pressure sensitive adhesive (PSA) composition. The PSA includes (a) at least one monomer which may undergo free radical polymerization and whose homopolymer has a glass transi- 50 tion temperature (Tg) of less than or equal to 10° C., (b) at least one mono-olefinically unsaturated monomer having aldehyde or ketone functionality, and (c) an effective amount of at least one polyhydrazide crosslinker having hydrazine functionality. The PSA composition of the invention exhibits 55 a peel adhesion value of between about 0.4 lbs/in (70N/in) and about 4 lbs/in (700 N/m), and the percent film remaining on the polymeric packaging after a sink-float testing is 0% to about 8%. In another embodiment, the PSA composition of the invention further includes a surfactant with a reactive 60 carbon-carbon double bond.

In another embodiment, there is provided a filmic faces-tock material for polymeric packaging which includes a PSA composition. The PSA composition includes (a) 50 wt % to about 95 wt % of at least one monomer which may undergo free radical polymerization and whose homopolymer has a glass transition temperature (Tg) of less than or equal to 10°

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C., (b) greater than 0.1 wt % at least one mono-olefinically unsaturated monomer having aldehyde or ketone functionality, (c) an effective amount of at least one polyhydrazide crosslinker having hydrazine functionality, and a surfactant with a reactive carbon-carbon double bond, with the wt % being based on the weight of the monomer mixture, with the effective amount of the polyhydrazide crosslinker is such that the ratio of the hydrazine functionality is between about 0.02 to 5 equivalents per 1 equivalent of the aldehyde or ketone functionality. The PSA composition of the invention exhibits a peel adhesion value of between about 0.4 lbs/in (70 N/m) and about 4 lbs/in (700 N/m), and the percent film remaining on the polymeric packaging after a sink-float testing is 0% to about 8%.

SUMMARY OF THE FIGURE

FIG. 1 is a scatter plot of Peel Adhesion v. Recyclability for PSA compositions of the invention and control PSA compositions.

DETAILED DESCRIPTION OF THE INVENTION

The removable PSA compositions of the invention includes an acrylic polymer dispersion containing a monomer mixture having (a) at least one monomer which may undergo free radical polymerization. In one embodiment, the monomer mixture may include at least one acrylic acid ester or (meth)acrylate acid ester whose homopolymer has a glass transition temperature (Tg) of less than or equal to 10° C., and (b) at least one mono-olefinically unsaturated monomer having an aldehyde or ketone group. The emulsion of the invention being reacted with (c) an effective amount of at least one polyhydrazide crosslinker.

Examples of the at least one acrylic acid ester or (meth) acrylate acid ester whose homopolymer has a glass transition temperature (Tg) of less than or equal to 10° C. include but are not limited to benzyl acrylate, n-butyl acrylate, sec-butyl acrylate, cyanoethyl acrylate, iso-decyl acrylate, n-decyl acrylate, dodecyl acrylate, dodecyl acrylate, 2-ethoxyethyl acrylate, ethyl acrylate, 2-ethylhexyl methacrylate, n-hexyl acrylate, hexyl methacrylate, isobutyl acrylate, isopropyl acrylate, isotactic, 2-methoxyethyl acrylate, methyl acrylate, octadecyl methacrylate, octyl methacrylate, 2-phenylethyl acrylate, n-propyl acrylate, and 2,2,2-trifluoroethyl acrylate, as well as mixtures or any subset thereof.

In one embodiment, Tg of the acrylic acid ester or (meth)acrylate acid ester homopolymer is between about -100° C. and 10° C., or between about -70° C. to 10° C., or between about -70° C. to -10° C.

In one embodiment, the at least one acrylic acid ester or (meth)acrylate acid ester whose homopolymer has a glass transition temperature (Tg) of equal to or less than 10° C. is present in an amount of about 50 wt % to about 95 wt % or about 70 wt % to about 90 wt % based on the weight of the monomer mixture.

In one embodiment, the at least one acrylic acid ester or (meth)acrylate acid ester whose homopolymer has a glass transition temperature (Tg) of equal to or less than 10° C. includes 2-ethylhexyl acrylate, n-butyl acrylate, or any mixtures thereof.

Mono-olefinically unsaturated monomers having ketone or aldehyde functionality are monomers containing an aldo or keto moiety and a polymerizable carbon-carbon double bond. Examples include, but are not limited to, acrolein,

methacrolein, vinylbenzaldehyde, crotonaldehyde, (meth) acryloxyalkylpropanals, diacetone acrylamide (DAAM), diacetone acrylate, or any mixtures or subsets thereof.

In one embodiment the amount of mono-olefinically unsaturated monomers having aldehyde or ketone function-5 ality is present in an amount of greater than 0.1 wt %, or about 0.5 to 10 wt. %, or about 0.75% to 5 wt. %, based on the weight of the monomer mixture.

In one embodiment, the mono-olefinically unsaturated monomers having ketone or aldehyde functionality includes diacetone acrylamide, diacetone acrylate, acrolein, or any mixtures thereof.

In one embodiment the monomer mixture may optionally include one or more acrylic acid ester or (meth)acrylate acid ester whose homopolymer has a Tg of greater than 10° C. 15 such as, for example greater than 10° C. to about 175° C. Examples include but are not limited to benzyl methacrylate, t-butyl acrylate, 2-tert-Butylaminoethyl methacrylate, butyl methacrylate, t-butyl methacrylate, cyclohexyl acrylate cyclohexyl methacrylate, diethylaminoethyl methacrylate, 20 dimethylaminoethyl methacrylate, ethyl methacrylate, hexadecyl acrylate, hexadecyl methacrylate, hexyl acrylate, 2-hydropropyl methacrylate, 2-hydroxyethyl methacrylate, isobornyl acrylate, isobornyl methacrylate, isobutyl methacrylate, isopropyl methacrylate, methyl methacrylate, atac- 25 tic, methyl methacrylate, syndiotactic, phenyl methacrylate and trimethylsilyl methacrylate, as well as mixtures or any subset thereof.

In one embodiment, the at least one acrylic acid ester or (meth)acrylate acid ester whose homopolymer has a glass 30 transition temperature (Tg) of greater than 10° C. is present in an amount of about 0 wt % to about 50 wt %, or about 0 wt % to about 30 wt % based on the weight of the monomer mixture.

In one embodiment, the monomer mixture may include 35 one or more ethylenically unsaturated carboxylic acid. Examples included but are not limited to monoethylenically unsaturated monocarboxylic acids such as acrylic acid, methacrylic acid, ethacrylic acid, crotonic acid, and carboxyethyl acrylate, monoethylenically unsaturated dicarboxylic acids such as maleic acid, fumaric acid, itaconic acid, and citraconic acid, monoethylenically unsaturated tricarboxylic acids such as aconitic acid, and the halogen substituted derivatives (e.g., alphachloracylic acid) and anhydrides of these acids (e.g., maleic anhydride and citraconic anhydride).

The PSA composition of the invention may comprise about 0.5 to about 7 wt % or about 1 to about 3 wt % ethylenically unsaturated carboxylic acids, based on the weight of the monomer mixture.

In one embodiment, the PSA composition of the invention may include an alkyl mercaptan chain transfer agent such as a $\rm C_6$ - $\rm C_{18}$ alkyl mercaptan for example n-hexyl mercaptan, n-octyl mercaptan and n-dodecyl mercaptan. When utilized, the PSA composition of the invention may comprise about 55 0.01 to about 0.5 wt % mercaptan chain transfer agents, based on the weight of the monomer mixture.

In another embodiment of the invention, the process to prepare the dispersions of the invention may utilize a continuous phase (usually water), and other components 60 standard and known in the art such as initiators, reducing agents, surfactants, catalysts, wetting agents, crosslinking agents, preservatives and the like. For example, any conventional water soluble polymerization initiator suitable for emulsion polymerization may be used. The typical wt % of 65 said initiators are from 0.01% to 2.0 wt. % and preferably 0.01 to about 1.0 wt % based on the total weight of

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monomers. Examples of initiators include, but are not limited to, persulfates, peroxides, azo compounds, and the mixtures thereof. The water soluable initiators can be used alone or in combination with one or more conventional reducing agents such as, but not limited to, sodium formal-dehyde sulfoxylate, sodium metabisulfite, ascorbic acid, ferrous salts, chelated iron salts, and the like.

In one embodiment, the water soluable polymerization intiator systems are peroxides utilized in combination with conventional reducing agents such as tert-butyl hydroperoxide sodium formaldehyde sulfoxylate based systems with a chelated ferric complex catalyst.

Examples of suitable surfactant systems are those known

in the art and include anionic, nonionic, cationic, or amphoteric emulsifiers and mixtures thereof. Examples of anionic surfactants include, but are not limited to, alkyl sulfates, sulfates of ethoxylate alcohols, aryl sulfonates, phosphates of ethoxylated alcohols, sulfosuccinates, sulfates and sulfonates of ethoxylated alkylphenols, and mixtures thereof. Examples of nonionic surfactants include, but are not limited to, ethoxylated alcohols, ethoxylated alkylphenols, and mixtures thereof. Examples of cationic surfactants include, but are not limited to, ethoxylated fatty amines. In one embodiment, reactive surfactant chemistry which have reactive carbon-carbon double bond is utilized. Examples of reactive chemistries include, but are not limited to, alkylphenol ethoxylates containing alkenyl substituents, polyoxyalkylene-1-(allyloxymethyl)alkyl ether sulfate salt compounds, salts of poly(oxy-1,2-ethanediyl), alpha-sunomega-[1-(hydroxymethyl)-2-(2-propen-yloxy)ethoxy] (Adeka SR Series, commercially available from Adeka Corporation), and mixtures or subsets thereof. In one embodiment, the reactive surfactant includes an Adeka SR Series reactive surfactant. The typical weight of surfactant is 0.2 to 5.0 wt. % and more preferably 1.0 to 5.0 wt. % and most preferably 1.0 to 3.0 wt. %. The surfactants are utilized by conventional methods that are well known in art. In one embodiment, the process to prepare the PSA includes the emulsification of the monomer mix with the surfactant system prior to the polymerization reaction.

Following polymerization, the pH of the latex emulsion is adjusted with a suitable base including, but are not limited to, metal hydroxides, ammonium hydroxide, amines, and mixtures thereof. The pH is adjusted to at least 6.0, and more preferably 6.5 to 9.5, and most preferably 7.0 to 9.0. In one embodiment, the suitable base is ammonium hydroxide.

The PSA emulsion of the invention has a typical solids content from about 30 to 70% and preferably 40 to 55%. Polymerization can be conducted at typical temperatures for emulsion polymerization. The polymerization is preferably conducted in the range of 110° F. (43.3° C.) to 210° F. (99° C.) and more preferably 130° F. (54° C.) to 190° F. (88° C.).

The PSA compositions of the invention are reacted with an effective amount of polyhydrazide compound having at least two hydrazide (NH—NH₂) functional groups. Specific examples of polyhydrazide compounds include, but are not limited to, oxalic acid dihydrazide, malonic acid dihydrazide, succinic acid dihydrazide, glutaric acid dihydrazide; adipic acid dihydrazide, sebacic acid dihydrazide; monoolefin unsaturated dicarboxylic acid dihydrazide such as maleic acid dihydrazide, fumaric acid dihydrazide, itaconic acid dihydrazide; carbonic acid dihydrazide such as carbonic acid dihydrazide; aromatic dihydrazide such as phthalic acid dihydrazide, terephthalic acid dihydrazide, isophthalic acid dihydrazide; trihydrazides such as 1,2,4-butanetricarbohydrazide, 1,1,4-butanetricarbohydrazide, 1,3,6-hexanetricarbo

hydrazide, 1,3,7-heptanetricarbohydrazide, and I-hydroxy-1,2,4-butanetricarbohydrazide, and mixtures or any subsets thereof. In one embodiment, the polyhydrazide includes adipic dihydrazide, isophthalic dihydrazide, sebacic dihydrazide, bis-semicarbizides, and mixtures thereof. In another embodiment, the polyhydrazide is adipic acid dihydrazide.

The effective amount of the polyhydrazide crosslinker is such that the ratio of hydrazine functionality is between about 0.02 to 5, or about 0.1 to 3 or about 0.5 to 2 equivalents per 1 equivalent of the ketone or aldehyde functionality of the mono-olefinically unsaturated monomers having ketone or aldehyde functionality. The polyhydrazide compound can be added at any point in the process. In one embodiment, it is added after the emulsion reaction. In one embodiment, the polyhydrazide crosslinker is optional.

The PSA compositions of the invention may be used with filmic or paper facestock materials. Examples of suitable filmic facestock materials include, but are not limited to, 20 olefinic facestocks (e.g. polypropylene, polyethylene, etc), polystyrene facestocks, PET facestocks, polyvinyl chloride and mixtures thereof. The facestock constructions can contain multiple layers. The facestock can also contain a top-coat, printing, and/or overlaminates.

In one embodiment the PSA compositions of the invention typically exhibit peel adhesion values of less than about 4 lbs/in (700 N/m), less than about 2 lbs/in (350 N/m), less than about 1 lb/in (175 N/m) or between about 0.4 lbs/in (70 N/m) to about 4 lbs/in (700 N/m) or between about 0.4 lbs/in (70 N/m) to about 2 lbs/in (350 N/m). In another embodiment, during the recycling of polymeric packaging with filmic labels adhered with the PSA compositions of the invention, the percent film (% film) remaining on PET after sink-float testing is about 0% to about 8%, or about 0% to about 5%, or about 0% to about 3%.

In one embodiment, the PSA compositions of the invention are free of additives as described in US Patent Publication No. 2010/0051200 as facilitating the caustic washing of the polymeric substrate during the recycling process. In is known in the art that use of such additives may impart moisture sensitivity to a PSA composition.

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In order to provide a better understanding of the present invention including representative advantages thereof, the following examples are offered.

EXAMPLES

Example 1

In a 2 L jacketed glass reactor equipped with a reflux condenser, thermocouple, and 4-blade agitator, 1800.0 g of latex was prepared. The reactor was charged with 410.7 g of water, 5.6 g of Abex 2535 surfactant (available from Rhodia Solvay Group), and 0.0040 g of Dissolvine 4.5 catalyst (available from Akzo Nobel N.V.), heated to 140° F. (60° C.), and agitated from 100 to 300 rpms. The monomer preemulsion was prepared in a separate vessel by mixing 257.7 g of water, 16.1 g of Adeka SR 3025 (available from Adeka Corporation) surfactant, 2.4 g of Abex 2535 surfactant, 8.05 g of Surfynol 440 (available from Air Products and Chemicals, Inc.) wetting agent, 12.1 g of betacarboxyethyl acrylate, 8.1 g acrylic acid, 40.3 g butyl acrylate, 40.3 g ethyl acrylate, and 724.8 g 2-ethylhexyl acrylate. After the preemulsion was mixed and formed, 32.8 g of preemulsion was added to the reactor. And additional 32.2 g of Adeka SR3025 was added to the preemulsion. 24.2 g of water with 0.4 g of tertbutyl hydroperoxide and 24.2 g of water with 0.2 g of sodium formaldehyde sulfoxylate was added to the reactor to create an in-situ emulsion seed. After the in-situ seed was formed indicated by an initial exotherm, the preemulsion was fed over 180 minutes and an oxidizer feed of 48.3 g of water with 2.4 g of tertbutyl hydroperoxide and a reducer feed of 96.6 g of water with 1.2 g of sodium formaldehyde sulfoxylate were added over 300 minutes. The reaction was carried out at 138° F. (59° C.) to 142° F. (142° C.). After 90 minutes of the preemulsion being fed, 24.2 g of a 30% diluted in water diacetone acrylamide solution was added to the preemulsion. When the oxidizer and reducer feeds completed, the reactor was cooled to <110° F. (43° C.) and 8.1 g of water with 6.4 g of 28% aqueous solution ammonium hydroxide was added. 4.0 g of water with 1.6 g of Acticide LA preservative (available from Thor Specialties, Inc.) was added to the reactor. Examples 2 to 10 were synthesized in the same manner as Example 1 with the changes to the compositions set forth in Tables 1 and 2.

TABLE 1

	Example					
	1 (Invention)	2 (Control)	3 (Control)	4 (Control)	5 (Invention)	
2-Ethylhexyl Acrylate	724.8	734.6	733.9	728.0	644.2	
Methyl Methacrylate	0.0	0.0	0.0	0.0	80.5	
DAAM 30% Solution	24.2	0.0	0.0	0.0	24.2	
n-Dodecyl Mercaptan	0.0	0.0	0.0	0.0	0.0	
Allyl Methacrylate	0.0	0.0	1.6	0.0	0.0	
Hexanediol Diacrylate	0.0	0.0	0.0	16.2	0.0	

TABLE 2

	Example					
	6 (Invention)	7 (Invention)	8 (Control)	9 (Invention)	10 (Control)	
2-Ethylhexyl Acrylate	604.0	721.5	731.3	563.7	733.8	
Methyl Methacrylate	120.8	0.0	0.0	161.1	0.0	
DAAM 30% Solution	24.2	24.2	0.0	24.2	0.0	
n-Dodecyl Mercaptan	0.0	0.0	0.0	0.0	0.8	
Allyl Methacrylate	0.0	3.2	3.3	0.0	0.0	
Hexanediol Diacrylate	0.0	0.0	0.0	0.0	0.0	

The pH of each example was adjusted with 26 Be ammonium hydroxide to a pH of 8.3 to 8.5, and 1.0% of Surfvnol 440 wetting agent was added. In the examples of the invention (i.e. examples 1, 5, 6, 7 and 9) a stoichomteric equivalent amount of adipic dihydrazide to diacetone acrylamide was added. Samples were coated onto paper coated with silicone release liner at 10 g/m² and dried for 1-minute at 90° C. Afterward, the samples were laminated to 1.6 mils cavitated white biaxial oriented polypropylene and accli- 10 mated at room temperature for greater than 24-hours. The samples were applied to the substrates and aged 1-hour at 50° C. followed by 1-hour cool to room temperature before

The examples were tested according to the small scale laboratory test as described in "APR Screening Plastic Label and Closure Floatability Test for PET Bottles" (see www-.plasticsrecycling.org). However, the ground piece size was reduced from 3/8 inch to 1/4 inch, which is believed to more 20 accurately represent flake found at the recycling plants. In addition, the test temperature was reduced to 85° C. from 88° C. Peel adhesion was tested according to the Pressure Sensitive Tape Council standard PSTC 101 on PET substrate (ASTM Designation D3330 Standard Test Method for Peel 25 Adhesion of Pressure-Sensitive Tape). The results are set forth in Table 3. The data in Table 3 is set forth in a scatter plot in FIG. 1.

testing.

TABLE 3

Example	Peel Adhesion lbs/in (N/m)	% Film Remaining on PET After Sink-Float
Example 1 Invention	0.532 (93)	1.0%
Example 2 Control	0.609 (106)	24.5%
Example 3 Control	0.484 (85)	8.5%
Example 4 Control	0.777(136)	37.0%
Example 5 Invention	0.697 (122)	1.0%
Example 6 Invention	0.774 (135)	0.0%
Example 7 Invention	0.38 (66)	0.0%
Example 8 Control	0.365 (64)	1.0%
Example 9 Invention	0.818 (143)	3.0%
Example 10 Control	1.29 (226)	64.0%

An APR approved lab, Plastics Forming Enterprise (PFE) (see www.plasticsforming.com), following APR protocol, 45 conducted an independent evaluation in the recycling process using the PSA composition of Example 1 and resulting PET recycled plaques, compared to control non-labeled PET plaques. The results for the L*a*b* color scale and haze are set forth in Table 4 [L* (100=white; 0=black) a* (positive=red; negative=green; 0=gray) (positive=yellow; negative=blue; 0=gray)]

Based on the above results, referring to the Examples of the invention, the addition of diacetone acrylamide chemistry to the PSA allows for recyclability at higher peel values which is desirable for labeling systems designed for polymeric packaging. In contrast, referring to control example 7, a peel level of below 0.40 lbs/in is needed to allow for sufficient label release from the PET flake during recycling.

We claim:

- 1. A filmic facestock material for polymeric packaging comprising:
 - a pressure sensitive adhesive composition comprising a monomer mixture, wherein the monomer mixture comprises (a) at least one monomer that is free radical polymerizable and whose homopolymer has a glass transition temperature of less than or equal to 10° C., (b) greater than 0.1 wt. % to 1 wt. % based on the weight of the monomer mixture of at least one monoolefinically unsaturated monomer having aldehyde or ketone functionality, and (c) at least one polyhydrazide crosslinker having hydrazine functionality;
 - wherein the pressure sensitive adhesive composition exhibits a peel adhesion value of between about 0.4 lbs/in (70 N/m) and about 4 lbs/in (700 N/m); and
 - wherein a percent film remaining on the polymeric packaging after a sink-float testing is 0% to about 8%.
- 2. The filmic facestock material of claim 1 wherein the at least one monomer that is free radical polymerizable and 30 whose homopolymer has a glass transition temperature of less than or equal to 10° C. comprises at least one acrylic acid ester or (meth)acrylate acid ester.
- 3. The filmic facestock material of claim 2 wherein the least one acrylic acid ester or (meth)acrylate acid ester is 35 selected from the group consisting of benzyl acrylate, n-butyl acrylate, sec-butyl acrylate, cyanoethyl acrylate, isodecyl acrylate, n-decyl acrylate, dodecyl acrylate, dodecyl methacrylate, 2-ethoxyethyl acrylate, ethyl acrylate, 2-ethylhexyl acrylate, 2-ethylhexyl methacrylate, n-hexyl acrylate, hexyl methacrylate, isobutyl acrylate, isopropyl acrylate, isotactic, 2-methoxyethyl acrylate, methyl acrylate, octadecyl methacrylate, octyl methacrylate, 2-phenylethyl acrylate, n-propyl acrylate, and 2,2,2-trifluoroethyl acrylate, and combinations thereof.
 - 4. The filmic facestock material of claim 3 wherein the least one acrylic acid ester or (meth)acrylate acid ester is selected from the group consisting of 2-ethylhexyl acrylate, n-butyl acrylate, and combinations thereof.
 - 5. The filmic facestock material of claim 1 wherein the at least one at least one mono-olefinically unsaturated monomer having an aldehyde or ketone functionality is selected from the group consisting of acrolein, methacrolein, vinyl-

TABLE 4

Example	L* Values	a* Values	b* Values	L* average	a* Average	b* Average	Haze	Haze Average
Example 1 Control PET	87.71 87.76 87.74 87.79 87.77 87.76 87.85	-1.16 -1.17 -1.16 -1.12 -1.13 -1.23 -1.21	3.68 3.82 3.61 3.34 3.43 3.74 3.68	87.75 87.78	-1.15 -1.21	3.58	2.67 2.65 2.59 2.42 2.48 2.47 2.17	2.56
	87.78 87.83 87.67	-1.21 -1.19 -1.22	3.63 3.5 3.76				2.57 2.08 2.96	

benzaldehyde, crotonaldehyde, (meth)acryloxyalkylpropanals, diacetone acrylamide, diacetone acrylate, and combinations thereof.

- 6. The filmic facestock material of claim 5 wherein the at least one at least one mono-olefinically unsaturated monomer having an aldehyde or ketone functionality is selected from the group consisting of diacetone acrylamide, diacetone acrylate, acrolein, and combinations thereof.
- 7. The filmic facestock material of claim 1 wherein the monomer mixture comprises: 50 wt % to about 95 wt % of the at least one monomer which may undergo free radical polymerization and whose homopolymer has a glass transition temperature of less than or equal to 10° C., and greater than 0.1 wt. % to 1 wt. % of the at least one monoolefinically unsaturated monomer having an aldehyde or ketone group, wherein the wt % is based on the weight of the monomer mixture.
- **8**. The filmic facestock material of claim **1** wherein the monomer mixture further comprises one or more ethylenically unsaturated carboxylic acid.
- 9. The filmic facestock material of claim 1 wherein the monomer mixture further comprises at least one monomer that is free radical polymerizable and whose homopolymer has a glass transition temperature of greater than 10° C.
- 10. The filmic facestock material of claim 1 wherein the hydrazine functionality is between about 0.02 to 5 equivalents per 1 equivalent of the aldehyde or ketone functionality.
- $\dot{1}$ 1. The filmic facestock material of claim 1 wherein the $_{30}$ polyhydrazide is selected from the group consisting of oxalic acid dihydrazide, malonic acid dihydrazide, succinic acid dihydrazide, glutaric acid dihydrazide; adipic acid dihydrazide, sebacic acid dihydrazide; monoolefin unsaturated dicarboxylic acid dihydrazide such as maleic acid 35 dihydrazide, fumaric acid dihydrazide, itaconic acid dihydrazide; carbonic acid 10 polyhydrazide such as carbonic acid dihydrazide; aromatic dihydrazides such as phthalic acid dihydrazide, terephthalic acid dihydrazide, isophthalic acid dihydrazide; trihydrazides such as 1,2,4-butanetricarbohydrazide, 1,1,4-butanetricarbohydrazide,1,2,5-pentanetricarbohydrazide, 1,3,6-hexanetricarbohydrazide, 1,3,7-heptanetricarbohydrazide, and I-hydroxy-1,2,4butanetricarbohydrazide, and combinations thereof.
- 12. The filmic facestock material of claim 11 wherein 45 polyhydrazide is selected from the group consisting of adipic dihydrazide, isophthalic dihydrazide, sebacic dihydrazide, bis-semicarbizides, and combinations thereof.
- 13. The filmic facestock material of claim 1 wherein the polymeric packaging comprises polyethylene teraphthalate, high density polyethylene, polyvinyl chloride, polypropylene or combinations thereof.
- **14**. The filmic facestock material of claim **1** wherein the filmic facestock comprises an olefinic facestock, a polystyrene facestock, a polyethylene teraphthalate facestocks, a polyvinyl chloride or combinations thereof.
- **15**. The filmic facestock material of claim **1** wherein the pressure sensitive adhesive composition further comprises a surfactant with a reactive carbon-carbon double bond.

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- 16. The filmic facestock material of claim 15 wherein the surfactant with a reactive carbon-carbon double bond is selected from the group consisting of alkylphenol ethoxylates containing alkenyl substituents, polyoxyalkylene-1-(allyloxymethyl) alkyl ether sulfate salt compounds, salts of poly(oxy-1,2-ethanediyl), alpha-sulfo-omega-[1-(hydroxymethyl)-2-(2-propen-yloxy)ethoxy] and combinations thereof.
- 17. The filmic facestock material of claim 1 wherein the facestock material is an olefinic facestock.
- 18. A filmic facestock material for polymeric packaging comprising:
 - a pressure sensitive adhesive composition comprising a monomer mixture, wherein the monomer mixture comprises (a) at least one monomer that is free radical polymerizable and whose homopolymer has a glass transition temperature of less than or equal to 10° C., (b) greater than 0.1 wt. % to 1 wt. % based on the weight of the monomer mixture of at least one monoolefinically unsaturated monomer having aldehyde or ketone functionality, and (c) at least one polyhydrazide crosslinker having hydrazine functionality, and a surfactant with a reactive carbon-carbon double bond;
 - wherein the pressure sensitive adhesive composition exhibits a peel adhesion value of between about 0.4 lbs/in (70 N/m) and about 4 lbs/in (700 N/m); and
 - wherein a percent film remaining on the polymeric packaging after a sink-float testing is 0% to about 8%.
- 19. A filmic facestock material for polymeric packaging comprising:
 - a pressure sensitive adhesive composition comprising a monomer mixture, wherein the monomer mixture comprises (a) 50 wt % to about 95 wt % of at least one monomer that is free radical polymerizable and whose homopolymer has a glass transition temperature of less than or equal to 10° C., (b) greater than 0.1 wt. % to 1 wt % at least one mono-olefinically unsaturated monomer having aldehyde or ketone functionality, (c) at least one polyhydrazide crosslinker having hydrazine functionality, and a surfactant with a reactive carbon-carbon double bond, where the wt % is based on the weight of the monomer mixture;
 - wherein the hydrazine functionality is between about 0.02 to 5 equivalents per 1 equivalent of the aldehyde or ketone functionality;
 - wherein the pressure sensitive adhesive composition exhibits a peel adhesion value of between about 0.4 lbs/in (70 N/m) and about 4 lbs/in (700 N/m); and
 - wherein a percent film remaining on the polymeric packaging after a sink-float testing is 0% to about 8%.
- 20. The filmic facestock material of claim 19 wherein the surfactant with a reactive carbon-carbon double bond is selected from the group consisting of alkylphenol ethoxylates containing alkenyl substituents, polyoxyalkylene-1-(allyloxymethyl) alkyl ether sulfate salt compounds, salts of poly(oxy-1,2-ethanediyl), alpha-sulfo-omega-[1-(hydroxymethyl)-2-(2-propen-yloxy)ethoxy] and combinations thereof.

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